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ON THE DETERMINATION OF DISLOCATION DENSITIES

by

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INTRODUCTION

Ever since experimental techniques for observing dislocations have been extant, one of the most commonly measured parameters in the study of dislocations has been the density of dislocations. Indeed whenever the yield strength of a material is mainly dependent on interactions between dislocations, one generally expects that the higher the density of dislocations, the higher will be the yield stress. Flow stress, electrical resistivity, work hardening rate, and many other parameters have been correlated with dislocation density in numerous investigations.

One of the problems in making an experimental determination of dislocation density by a direct observation technique is the lack of a spatially continuous distribution of dislocations throughout a material. Dislocations cluster near grain boundaries, form "tangles", and generally do not arrange themselves in a uniform manner. This is the main problem that will be examined here. A discussion of problems in making measurements of density in non-homogeneous media will first be given.

SECTION I.

DENSITY DETERMINATIONS FOR NON-HOMOGENEOUS MEDIA

When one is measuring the mass density of a material like steel, one simply selects a piece of it with a shape which has an easily determined volume, say a cube, and with a mass which is readily determined by a convenient scale or balance. Since the material is macroscopically homogeneous it matters little if the piece has a volume of one cubic centimeter or one cubic meter; one will obtain the same mass density. However, for certain materials, namely non-homogeneous materials, the size of the object one uses to make density determinations does matter.

The determination of mass density for a material such as sintered WC-Co could be uncertain if it were possible to get a small enough volume so that only a very small number (or no) WC particles were in the Co matrix (or if only one WC particle were used with no Co). On an even smaller level of measurement, if, in the Co, one could determine the mass of a volume containing only a few atoms, one would again have an uncertainty in the determination of mass density. Long (1961) gives a discussion similar to that given here for the case of fluids.

SECTION II.

RELATIONSHIPS BETWEEN VARIOUS DISLOCATION DENSITY DEFINITIONS

Dislocations are considered as line elements in a volume, so the definition of dislocation density which is normally used is the line length of dislocations per unit volume. This is the "true" or "length" density, ρ_L, and its dimensions are $[L^{-2}]$. Another density definition, which is employed in surface investigations of dislocations such as the etch-pit method, is the "intersection" density, ρ_I. The intersection density is the number of dislocation lines piercing a unit surface. The dimensions of intersection density are also $[L^{-2}]$. The question arises as to whether these two densities are numerically equal.

Hirsch (1956) and Lomer (1959) state that the total line length in a unit volume is three times the number piercing a unit area. However, Frank (1957) and Livingston (1962) point out that Hirsch and Lomer were wrong and that the factor should be two. An example which shows the probable source of Hirsch and Lomer's error follows. An appealing dislocation configuration to take is straight dislocations running parallel to the edges of a unit cube. If we take 100 dislocations equally distributed over each face and one cm as the edge length, we have a total line length of dislocations equal to 300 cm. The length density is then 300 cm^{-2}. If we count the points of emergence in one plane and divide by the area, we get the intersection density. Table 1 shows the ratio of length and intersection densities when using different "counting planes" (Figure 1). We see that by taking different counting planes we obtain different values of ρ_I and thus different values of ρ_L/ρ_I. The problem with this approach is that the dislocation lines are not randomly arranged. This is such an

appealing configuration for obtaining a correlation between intersections and dislocations (and dislocation lengths) that as late as 1964, Akulov (1964, p770) uses it also and incorrectly relates intersections with total number of dislocations.

Since only indications about how to derive a relationship between ρ_I and ρ_L are given in the literature (Frank, 1957; Livingston, 1962), a rigorous derivation is given here. In figure 2a, a unit cube is shown with all dislocations oriented at an angle ϕ to the normal to the horizontal faces. All are drawn in the same direction for clarity. The intersection density on the top face is the number of intersections, N, divided by unit area, so $\rho_I = N$.

The length density is (N)(L) divided by unit volume. Thus $\rho_L = N/\cos\phi$. Hence $\rho_I = \rho_L \cos\phi$. This is only for one angle, ϕ. If we now take the spatial average ρ_I over the volume of the unit sphere, figure 2b, we get:

$$\rho_I = \frac{\int_V \rho_L |\cos\phi|\, dV}{\int_V dV}$$

where dV is the differential volume $dV = r \sin\phi\, d\Theta\, d\phi\, dr$

$$\rho_I = \frac{2\rho_L \int_{\phi=0}^{\pi/2} \cos\phi \sin\phi d\phi \int_{\Theta=0}^{2\pi} d\Theta \int_{r=0}^{1} rdr}{\int_0^{\pi} \sin\phi\, d\phi \int_0^{2\pi} d\Theta \int_0^1 rdr}$$

$$\rho_I = \frac{2\rho_L}{4\pi} \cdot 2\pi \left. \frac{\sin^2\phi}{2} \right|_0^{\pi/2}$$

$$2\rho_I = \rho_L \qquad (1)$$

In transmission electorn microscopy, one can measure the total projected length of dislocation lines, ℓ', in a given area. Since the projected length, L', of a dislocation of length L is $L|\sin\phi|$ (see figure 2a), we can do the same analysis as we did in the preceeding paragraph to obtain a relationship between true, ℓ, and projected total dislocation length for a random distribution:

$$\ell' = \Sigma L' = \frac{\int_V \Sigma L|\sin\phi|dV}{\int_V dV} = \frac{\ell \int_0^{\pi} \sin^2\phi d\phi \int_0^{2\pi} d\Theta \int_0^1 rdr}{\int_0^{\pi} \sin\phi d\phi \int_0^{2\pi} d\Theta \int_0^1 rdr}$$

$$\ell' = \frac{2\ell\pi}{4\pi} \int_{\phi=0}^{\pi} \sin^2\phi d\phi = \frac{\ell}{4} \int_0^{\pi} (1-\cos 2\phi)d\phi$$

$$\ell' = \frac{\pi}{4} \ell \qquad (2)$$

Equation (2) has been indicated previously by Bailey and Hirsch (1960).

Knowing the thickness of the specimen, t, and the area over which the projected length, ℓ', was determined, a value of ρ_L can be found from equation (2):

$$\rho_L = \frac{4}{\pi} \frac{\ell'}{At} \qquad (3)$$

Although this is a valid method, it certainly is very tedious to use since the measurement of ℓ' is very time consuming for the large areas which are required since dislocations are not uniformly distributed throughout the material.

To shorten the time consumed in estimating dislocation densities, Ham (1961) applied the Buffon "needle" problem as extended by Smith and Guttman (1953). Instead of measuring the projected length of dislocation lines per unit area, the number of intersections, n, of dislocations with a set of random grid lines of total length R can be counted. Using Smith and Guttman's equation:

$$\frac{\ell'}{A} = \frac{\pi}{2}\frac{n}{R} , \tag{4}$$

Ham modified equation (3) to find:

$$\rho_L = \frac{2n}{Rt} \tag{5}$$

provided n is large enough. In Ham's severely work hardened aluminum, using five lines drawn in random directions on a picture taken at 20,000X gave a large enough value of n ($\sim$ 50).

Equations (1) through (5) all depend upon a spatially random orientation of dislocation lines. The relationship between ρ_I and ρ_L, equation (1), also assumes that each dislocation piercing the surface is indicated (e.g., by an etch-pit). In addition to the requirement of randomness, an assumption made in obtaining equation (2), and thus equations (3) through (5), is that all dislocations are visible in the projected image.

SECTION III

THE VALIDITY OF ASSUMPTIONS MADE IN THE DERIVATION OF DISLOCATION DENSITY RELATIONSHIPS

Dislocations are seldom found to be uniformly distributed throughout a specimen. They tend to form in tangles and networks, thus causing non-uniform dislocation densities in a specimen (Wilsdorf, 1963). Also, Prince and Richman (1969) in their study of Al-Si alloys have shown that the smaller the silicon particle in the aluminum matrix, the larger the relative dislocation density in the vicinity of the particle.

In the etch-pit method of observing dislocations an etchant preferentially attacks the regions where dislocations (and other areas of atomic mismatch) pierce the surface causing etch-pits. The longer the etchant is allowed to stay in contact with the material, the larger the etch-pits. For optimum resolution, etch-pits should be as small as possible while still being discernable. To increase the resolution, Livingston (1962) and others have used shorter etching times which gave optically non-discernable etch-pits but these can be seen by electron microscopy by using a replication technique. The resolution limit for optical microscopy is about 0.7μ for the determination of etch-pits ($\rho \sim 10^8\ cm^{-2}$) while that for the replication technique is about 0.2μ ($\rho \sim 10^9\ cm^{-2}$). The spacing in very high density regions can be less than this, so etch-pit methods generally underestimate dislocation densities.

The question of whether there is an etch-pit for every dislocation and a dislocation for every etch-pit has not been resolved for all materials. Dash's (1957) work of decorating dislocations in silicon using copper nitrate has shown a definite one-to-one correspondence of etch-pits to decorated dislocations. Gilman and Johnston (1957) have convincingly

argued for one-to-one correspondence in LiF crystals. One of their strongest points is that when dislocations move away from original emergence points, subsequent etching reveals flat bottomed pits. Measured diffraction data for X-ray investigation of LiF is also pointed out by them as evidence for one-to-one correlation since it relates closely to the number of etch-pits formed.

Livingston (1962) points out the consistency of his data (etch-pits in Cu on [111] planes) and electron microscopic investigations by Bailey. However, Bailey (1963) rightly feels that this agreement is fortuitous since electron microscope techniques also underestimate dislocation density as we will see below.

Another investigation which examines the question of one-to-one correspondence is the work of Ruff (1962). In Cu he etched [111] planes of 1000 Å thick electron microscope specimens. In each case Ruff observes more than one dislocation per etch pit, but also notes that some pits are caused by impurity aggregates which tends to make the etch-pit density closer to the true ρ_L. Unfortunately, after etching, the very thin foils had to be washed, dried, and sometimes remounted for electron microscope investigation. This process makes it possible for new dislocations to be introduced and for old ones to move. Ruff took great pains to minimize this and feels that his findings are not greatly in error.

The resolution of the Berg-Barrett (reflection) and Lang (transmission) topographic methods of X-ray microscopy limits their usefulness to dislocation densities less than about 10^5 cm^{-2} (Otte and Hren, 1966). The Debye-Scherrer X-ray diffraction method will permit

information to be gained about dislocation densities and distribution up to the highest obtainable densities but with less detail (Gay et al, 1953). Hordon (1962) states that "...errors in the X-ray measurements due to instrumental broadening tend to overestimate dislocation density..."

In electron microscopy, two major factors limit the accuracy of dislocation density determinations. The rearrangement of dislocations during thinning and the overlapping of dislocation images both affect experimental ρ_L values.

In an early study, Ham and Sharpe (1961) argue that if the dislocations in thin foils are really random, the intersection density as obtained by counting the ends of lines on an electron-micrograph should equal that obtained by the random lime method, equation (5). They found that in cold-worked aluminum the intersection density was 20% greater than the density found by the random line method. This indicates that in the thinned foil the dislocations are preferentially oriented toward the normal to the surface. Later, Valdrè and Hirsch (1963) observed in 18-8 stainless steel ($\sim$ 3000 Å thick film) that during electropolishing, about 20% of the dislocations move. The general character of the networks, etc., are not changed, but the dislocations move to relieve internal stresses and shorten by rotating in their slip planes. The translation of dislocations does not alter accuracy of estimates of ρ_L, but rotations do alter the accuracy since there is then preferential orientation. They estimate that in networks their value of ρ_L may be in error in thin foils by as much as 30%. Dislocation loops near the surface may slip out of the foil due to image forces (Hirsch and Schmitz, 1962). This is a case where translations of dislocations does matter in deference to Hirsch's own previous statements.

In electron microscopy investigations, particularly in thicker foils, the overlapping of dislocation images becomes serious. As the foils are made thinner, overlapping is less of a problem but the dislocations adjust their positions so the rearrangement error becomes greater (Wilsdorf, 1963). Also, care must be taken so that all images are visible. This can be checked by tilting the specimen (Otte and Hren, 1966).

As has been pointed out by Seeger (1964), the significance of counting mean, as opposed to local, dislocation densities in all methods is questionable. Such mean densities underestimate the importance of the regions of low dislocation density where most of the dislocation movement during plastic deformation may occur. Also, the addition of more dislocations to an already "impenetrable" barrier will increase the dislocation density but will not change the effect of the barrier.

CONCLUSIONS

As has been rigorously derived here, the "true" or line length dislocation density is twice the "intersection" density. The relationships between these densities and various geometrical parameters have been derived and their applicability discussed. In the case of dislocation density determinations in very thin foils using transmission electron microscopy, the assumption of a random distribution of dislocation lines is not physically reasonable.

Further work needs to be done to put the thin foil dislocation density determinations on a more sound physical footing. This further work should include quantitative estimates (from geometrical arguments) of the change in dislocation density due to dislocations rearranging themselves to at least partially relieve internal stresses.

It may be convenient to argue that dislocation densities need not be known too accurately, so why bother with "exact" mathematical analyses? The obvious answer is that if you can do something correctly with little (or no) extra work, why not?

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TABLE I

COMPARISON OF LENGTH AND INTERSECTION DENSITIES FOR A NON-RANDOM DISLOCATION ARRANGEMENT (LINES PARALLEL TO THE THREE COORDINATE AXES) ($\rho_L = 300\ cm^{-2}$)

Plane	Emergence Points	Area (cm^2)	ρ_I (cm^{-2})	$\frac{\rho_L}{\rho_I}$
(100)	100	1	100	3
(110)	100 x 2	$\sqrt{2}$	142	2.1
(111)	50 x 3	$\frac{\sqrt{2}}{3}$	173	1.7

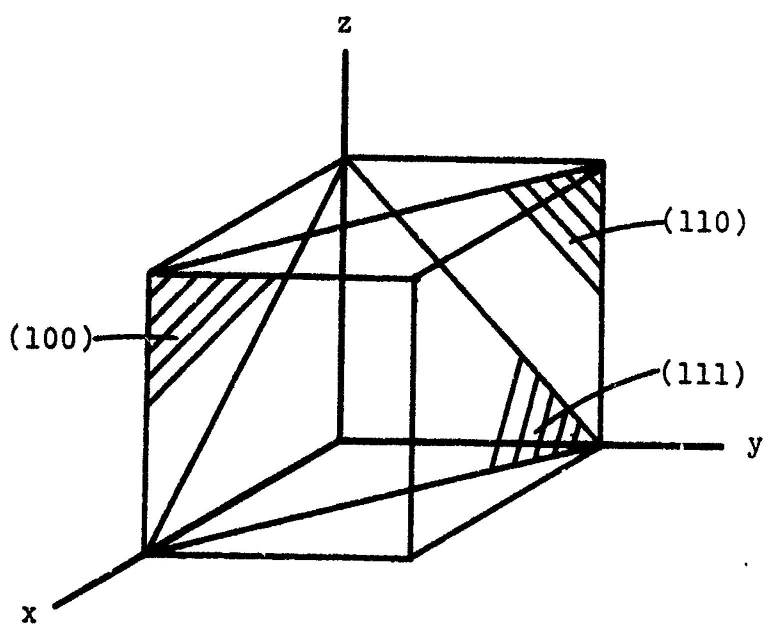
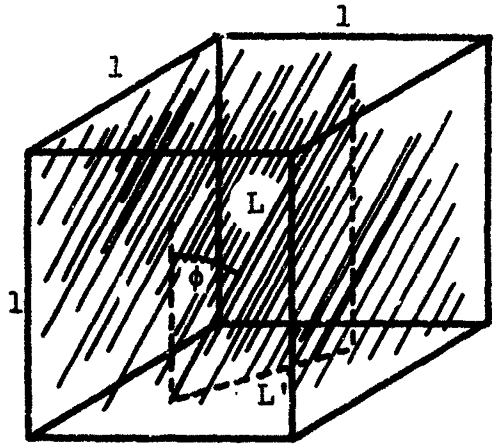


FIGURE 1 "Counting Planes" on unit cube



$$\ell' = \Sigma\, L'$$

$$\ell = \Sigma\, L$$

$$\rho_I = \frac{N}{(1)^2}$$

$$\rho_L = \frac{N \cdot L}{(1)^3} = N \cdot \frac{1}{\cos\phi}$$

$$\rho_I = |\cos\phi|\, \rho_L$$

FIGURE 2a. Relationship between ρ_I & ρ_L for arbitrary ϕ.

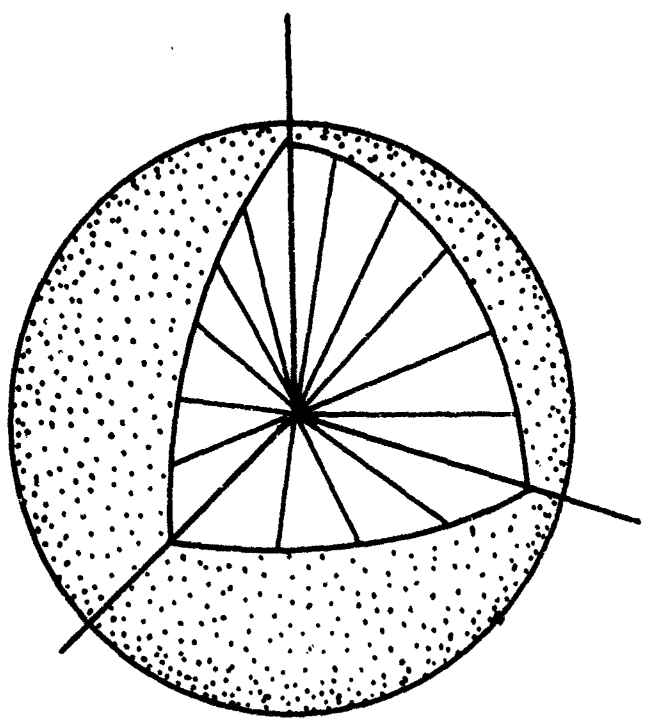


FIGURE 2b. Unit Sphere with spatial average of dislocation directions.

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